

## COLUMN AMOUNTS OF TRACE GASES FROM GROUND BASED FTIR MEASUREMENTS IN THE LATE NORTH POLAR WINTERS 1990 AND 1991

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### ABSTRACT

Two FTIR spectrometers were employed in the late winters 1990 and 1991 in Esrange, North Sweden, and in Ny Aalesund, Spitsbergen to detect zenith column amounts of several trace gases. Time series of column amounts of the trace gases O<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub>, HNO<sub>3</sub>, NO<sub>2</sub>, HCl, and HF have been derived from the measured spectra. Additionally, some information on the vertical distribution of HCl could be obtained by analyzing the spectral line shapes. The results are interpreted in terms of dynamical and chemical processes.

### 1. EXPERIMENTAL

The field campaigns were performed in the late winters 1990 and 1991 in Esrange, North Sweden (68° N, 21° E), from January 27 to March 31, 1990 and from January 15 to February 17, 1991, and in Ny Aalesund, Spitsbergen (79° N, 12° E), from March 13 to April 24, 1990. In Esrange, the uncooled laboratory version of the Fourier transform infrared spectrometer MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) was employed. In Ny Aalesund we used a modified BOMEM-DA2 spectrometer. The spectrometers have an unapodised spectral resolution of 0.015 cm<sup>-1</sup> and 0.010 cm<sup>-1</sup>, respectively.

### 2. DATA ANALYSIS

Interferograms were recorded on days with appropriate weather conditions by co-adding scans during an integration time of less than 15 minutes. From the interferograms, spectra were calculated by correcting the phase error and using a standard Fourier transformation. They were evaluated by fit-

ting simulated spectra, calculated using the SCAIS level-by-level and line-by-line algorithm, to the measured ones by scaling first guess mixing ratio profiles of the investigated gases. Model atmospheres for each day of measurement were generated from radiosonde data and ozone soundings for the simulations. Finally, the gas amounts along the vertical path were integrated from the scaled profiles.

Furthermore, some low resolved information on the vertical distribution of N<sub>2</sub>O and HCl could be obtained from a line shape analysis. A more detailed description of the experiment and the analysis procedure can be found in (Adrian et al., 1991) and (Adrian et al., 1992).

### 3. RESULTS AND DISCUSSION

From the spectra taken in Esrange in the two winters, time series of zenith column amounts of the gases O<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub>, HNO<sub>3</sub>, NO<sub>2</sub>, HCl, and HF have been derived. They are presented in Fig. 1 and Fig. 2, together with the stratospheric minimum temperatures taken from radiosonde data. In Ny Aalesund, spectra could be recorded only in the short period from March 24 to April 6, 1990, due to inappropriate weather conditions. Zenith column amounts for April 4, 1990 are given in Table 1.

Both the derived integrated zenith column amounts and the low resolved vertical distributions may be interpreted in terms of dynamical as well as chemical processes. During the late winter 1990, periods of strongly perturbed stratospheric chemistry have been identified. Our results indicate evident similarities to ground based Antarctic measurements reported by other groups. Distinct differences regarding the behaviour of the HNO<sub>3</sub> column amounts were found, which were much higher than

O <sub>3</sub>	HCl	HCl <sub>strat</sub>	HF	NO <sub>2</sub>	N <sub>2</sub> O	CH <sub>4</sub>
7.1 × 10 <sup>18</sup> ± 12 %	3.2 × 10 <sup>15</sup> ± 12 %	2.5 × 10 <sup>15</sup> ± 12 %	1.9 × 10 <sup>15</sup> ± 14 %	7.9 × 10 <sup>15</sup> ± 30 %	5.4 × 10 <sup>18</sup> ± 6 %	3.5 × 10 <sup>19</sup> ± 12 %

Table 1: Zenith column amounts in molecules/cm<sup>2</sup> measured in Ny Aalesund, Spitsbergen, at April 4, 1990 for observation angles between 4.9° and 16.7°. The results from different analyzed lines and observation angles were averaged.

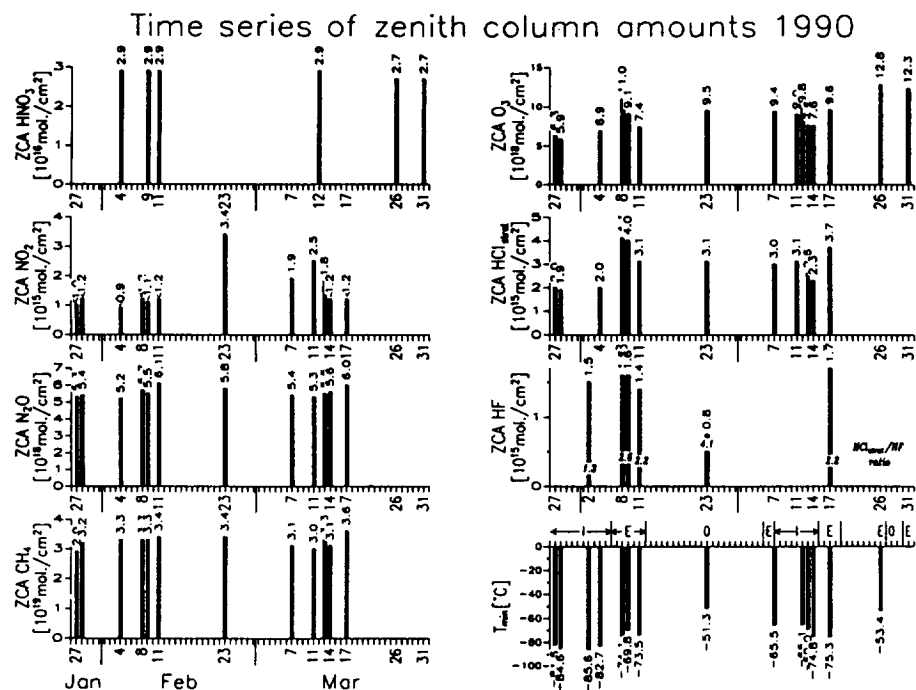


Fig. 1: Time series of zenith column amounts (ZCA) measured in Esrange from January 27 to March 31, 1990, and the stratospheric minimum temperatures taken from radiosoundings. The symbols I, E and O mark the periods when Esrange was inside, at the edge and outside of the polar vortex. For  $\text{NO}_2$ , values marked by '\*' represent upper limits of the zenith column amounts due to the detectability limit of the measurement.

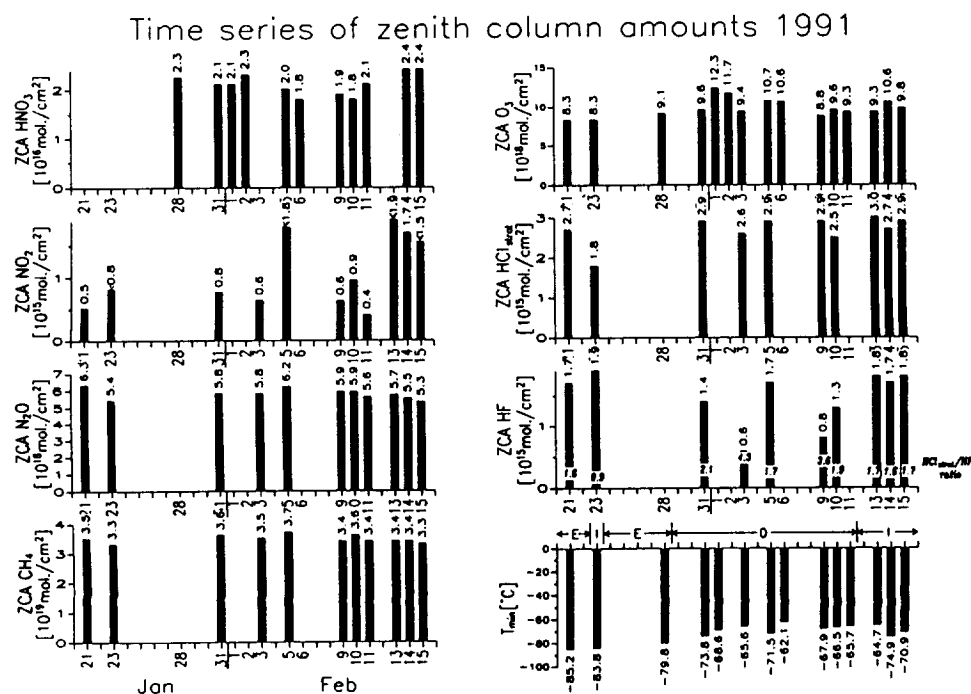


Fig. 2: Same as Figure 1 for the period from January 21 to February 15, 1991.

reported for Antarctica (e.g. Farmer et al., 1987), but consistent to aircraft measurements performed in the Arctic winter 1989 (Mankin et al., 1990).

Of particular interest is the increase of the HCl column amount from February 4 to 8, 1990 by nearly a factor of two, due to a strong increase in the lower stratosphere (see Fig. 3). Further understanding of the stratospheric situation is provided by an investigation of the dynamical and temperature history of the airmasses sampled with the help of a backward trajectory analysis (provided by the FU Berlin) (Petzoldt et al., 1991), which is shown in Fig. 4 and Fig. 5. From the dynamical analysis, subsidence of stratospheric air can be excluded as reason for the strong HCl increase. The differences in the temperature history of the air masses arriving in Esrange at February 4 and 8, respectively, indicate chemical reconversion of HCl by a reaction of Cl with CH<sub>4</sub> in the absence of PSCs, or conservation of HCl in PSC-free air masses.

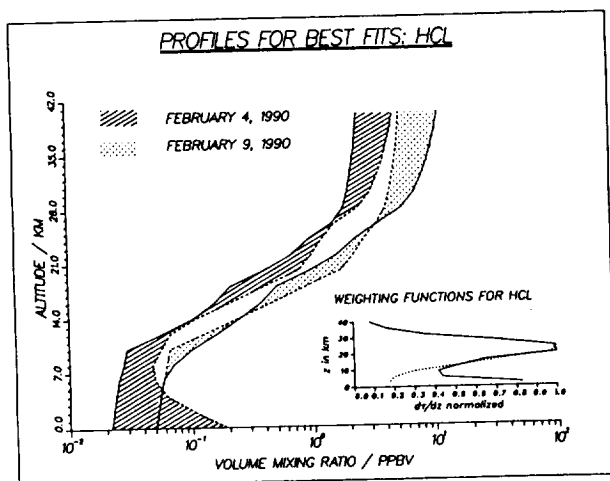


Fig. 3 Derived vertical concentration profiles and weighting functions of HCl for February 4 and 9, 1990. The shaded areas mark the range of uncertainty, in which the shapes of the measured HCl lines are fitted equally well. The weighting functions for HCl are calculated with the enveloping profiles derived for February 4. They describe the altitude region where the measurements are most sensitive to the trace gas concentration.

The late winters of 1990 and 1991 differ strongly in their meteorological situations. Whereas the stratospheric winter 1990 was very cold with a stable vortex until April, the winter 1991 can be characterized by a major midwinter warming (Naujokat et al., 1991).

During our observation period, Esrange was inside the polar vortex only at January 23, 1991 and from February 12 on, in the edge region of the vortex at January 21 and from January 24 to 28, and outside at the other observation days. In general, the results during this complicated stratospheric

situation are difficult to interpret without detailed dynamical analysis. The lowest O<sub>3</sub> column amounts during our observation period were found for the days inside the polar vortex. The low N<sub>2</sub>O, CH<sub>4</sub>, and HCl column amounts as well as the high HF burdens characterize the vortex air to be in a similar way chemically perturbed as found for 1990. In particular, the HCl/HF ratio is near 1.0 at January 23, which indicates conversion of HCl to other chlorine compounds due to heterogeneous chemical processes on PSCs.

At the beginning of February, the vortex broke down due to the major midwinter warming and reestablished in the second half of February. The expected vertical transports during the major midwinter warming are reflected in highly variable HF and NO<sub>2</sub> column amounts. Furthermore, for the tropospheric source gases N<sub>2</sub>O and CH<sub>4</sub> high column amounts comparable to midlatitude mean values are found.

The second half of February when the stratospheric circulation returned to late winter conditions can be characterized by relatively low O<sub>3</sub> columns, lowered N<sub>2</sub>O and CH<sub>4</sub> columns, and increased HF burdens. In comparison to the situation at the end of January, the NO<sub>2</sub> burdens are increased now by a factor of three, which still needs further understanding. During the whole observation period, the HNO<sub>3</sub> column amounts are lower by one third compared to the winter 1990, being highest in the middle of February and lowest during the major midwinter warming phase. Further interpretation of these results has to be done.

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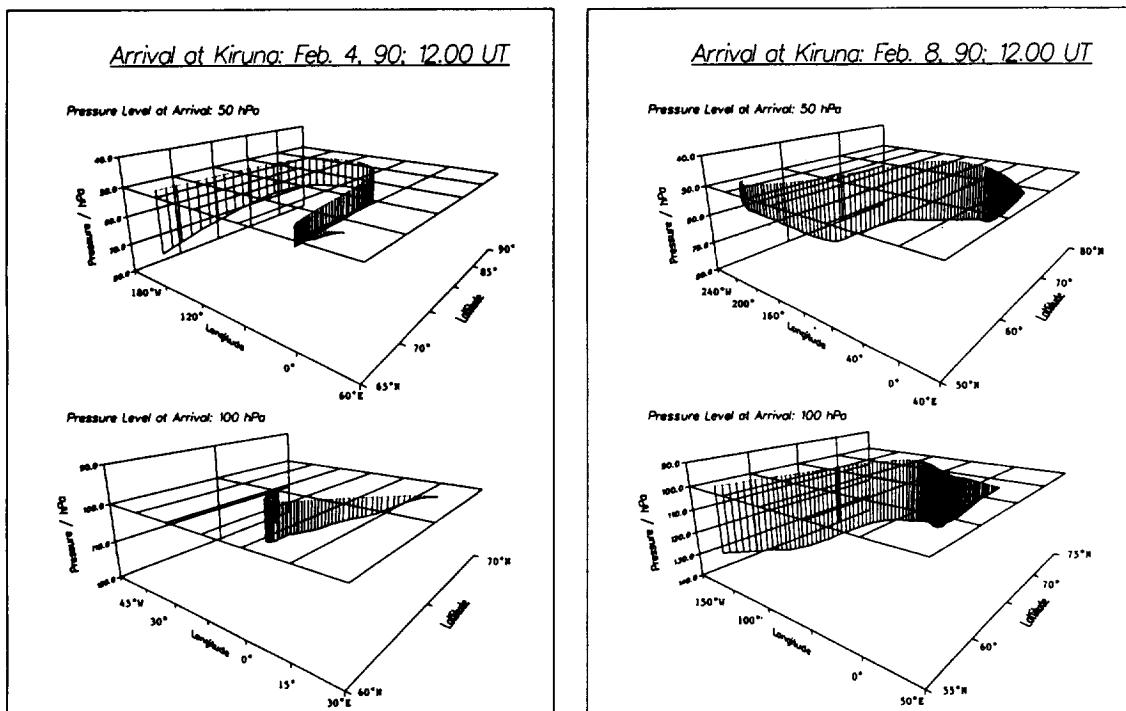


Figure 4: Pressure levels and geographic positions of air parcels along their trajectories for the arriving levels of 50 hPa and 100 hPa over Esrange at February 4 and 8, 1990. Time steps of one hour are marked by vertical lines. The upward motion of the air parcels along the trajectories of the 50 hPa arriving level exclude an increase of the HCl column amount due to dynamical reasons (compare Fig. 3).

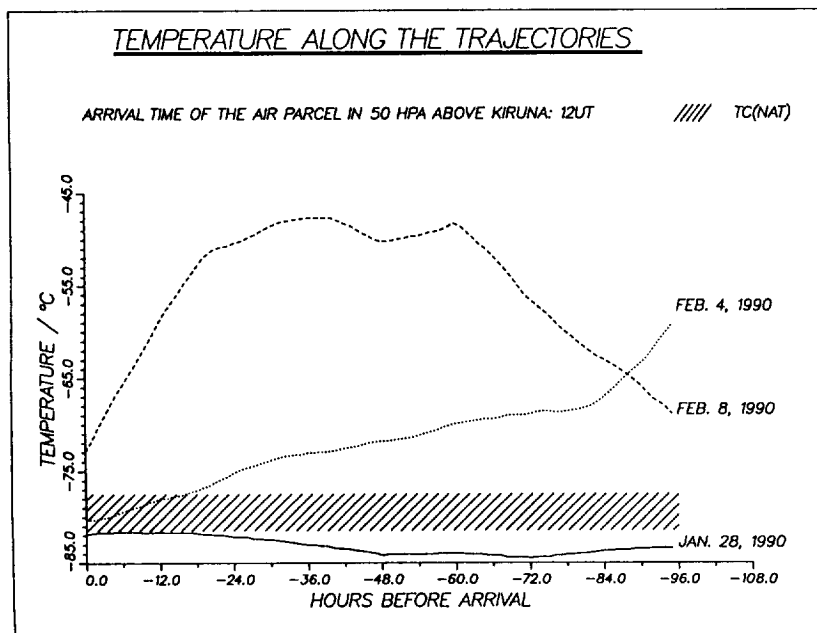


Fig. 5: Temperature history of an air parcel along its trajectory, arriving above Esrange in 50 hPa at 12 UT at January 28, February 4 and February 8, respectively (Petzoldt et al., 1991). The shaded area marks the range of the condensation temperature of NAT along the trajectories, considering a possible variability of the  $\text{HNO}_3$  concentration and the pressure dependence. The temperature histories for the air parcels arriving at January 28 and February 8, respectively, differ strongly, which may explain the strong increase of HCl between these dates.